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**T. Pollak  
Sanders Associates, Inc.  
95 Canal Street  
Nashua, New Hampshire 03061**

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**15 December 1980**

**Final Report  
For Period 27 September 1980 - 30 November 1980**

**Prepared for:  
U.S. Naval Reserach Laboratory  
4555 Overlook Avenue, S.W.  
Washington, DC 20375**

**Contract N00173-79-C-0343**

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Research and development on the crystalline laser host, YLF (LiYF <sub>4</sub> ), was completed during this program. The study involved crystal growth and sample fabrication of rare earth doped YLF. These materials were then evaluated at NRL. A total of 16 laser samples, eight different compositions, were processed during this contract period.		

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SECTION 1  
INTRODUCTION AND SUMMARY

The objective of this materials program was to perform crystal growth research and development for the laser host YLF ( $\text{LiYF}_4$ ). Crystalline boules of YLF, doped with various rare earth ions, were grown. Suitable laser specimens were then fabricated from this material. These laser specimens were then to be delivered to NRL with appropriate documentation.

Eleven growth attempts were made during this program, in which eight boules of various compositions were yielded. These boules were grown at two similar growth stations employing identical growth parameters. Crystal quality and yield improved in the later portion of the program as the result of equipment upgrade and the switch to a high grade argon gas atmosphere.

A total of 16 laser samples and rods were fabricated and delivered to NRL under this contract. These samples were cut and polished to standard laser specifications of flatness, parallelism, and surface finish.

A growth summary report was prepared for each set of laser samples.

## SECTION 2

### CRYSTAL GROWTH TECHNIQUE AND RESULTS

#### 2.1 TECHNIQUE

##### 2.1.1 INTRODUCTION

Sanders has developed techniques for automated growth of large, high quality crystals of the laser host  $\text{LiYF}_4$  and other fluorides with high yield. The basic approach chosen was the use of a computer-controlled process using a diameter sensor in a closed process loop. Close control of the diameter leads to higher quality material and reduction of material waste during fabrication.

The central item of Sanders' facility is a special-purpose crystal growing furnace designed on the experience obtained in the growth of laser materials at MIT and elsewhere. Growth of high quality crystals with high yield imposes three requirements:

- Uniform growth conditions
- Uniform, high purity feed
- Furnace integrity to prevent contamination during growth

Growth uniformity is assured by a closed-loop diameter control utilizing a novel electro-optic sensor and a digital computer. Figure 1 shows a view of the furnace and its control system.

Sanders has investigated a variety of feed purification techniques and have developed procedures to insure uniform quality starting materials irrespective of contaminants in the feed materials obtained from chemical companies.



Figure 1. Crystal Growth Furnace

## 2.1.2 TECHNIQUES

### 2.1.2.1 Growth Furnace

The growth furnace uses a vacuum technology for basic construction and seal design. This provides a system that can effectively prevent any interchange between the internal and external atmospheres for an extended period. When an inert atmosphere, such as helium or argon, is employed very high purity gases can be used without changing during the course of a run. The interior of the furnace is constructed with few areas that cannot be easily cleaned by reaching through the door opening. The entire growth chamber has water cooled walls to remove the radiated and conducted heat, and reduce the outgassing that would occur if the temperature of the walls were to increase. The upper chamber is uncooled because of the minor amount of heat that can reach the area.

An overall view of the growth furnace and its accessories is shown in Figure 1. It uses the double chamber principle as an interlock system for changing the seed without disturbing the melt in the lower chamber. A valve is used between chambers to permit the seed rod to pass through when the valve is open, yet separating the chambers when closed. The seed rod and the crucible positioning rod each pass through a double pumped seal to prevent inboard leakage that would contaminate the high purity atmosphere contained within the surface.

A graphite resistance heating element surrounds the outer surface of the platinum crucible in its graphite support; the heat zone is insulated by a heat shield made of six layers of molybdenum sheet. The heat shields are relatively poor insulation, but cause minimum contamination of the atmosphere. The heat load that arrives at the interior wall of the furnace chamber is removed by water flowing between the double walls of the lower chamber. Doors provide access to each of the chambers; the furnace with the doors open is shown in Figure 2 with the heat shield pack removed. Four windows are provided in the lower chamber for viewing the growth process.

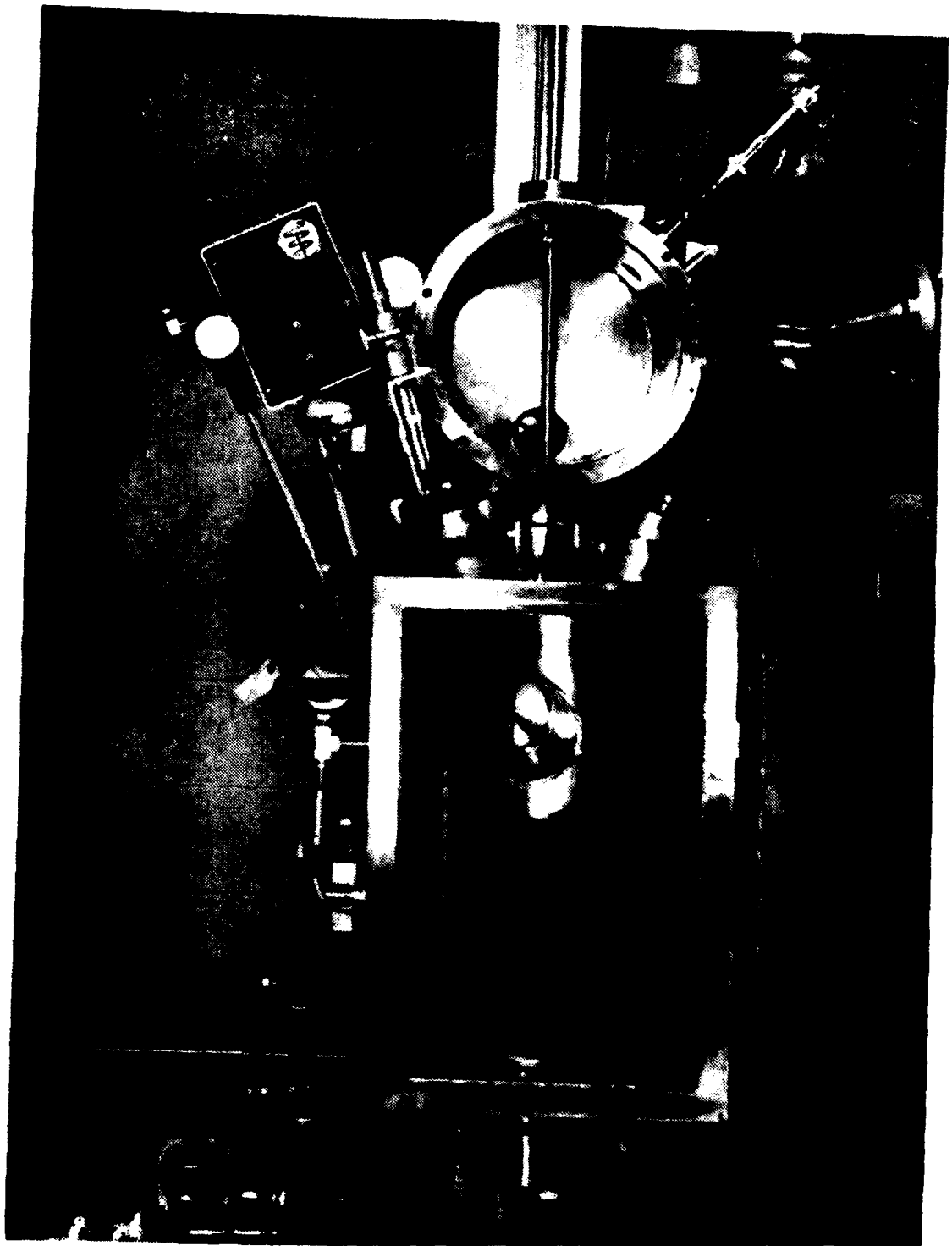


Figure 2. Crystal Growth Furnace Chamber

The furnace chambers can be pumped independently by a common high vacuum pumping system. Typical pressures that are attained after overnight pumping are about  $2 \times 10^{-7}$  Torr at the ionization gage.

#### 2.1.2.2 Controller

The control system philosophy was chosen to reduce required operator attention to a minimum by providing critical control loops that furnish the feedback that the operator would otherwise provide. Decreasing the required operator attention has a strong advantage beyond the obvious cost reduction. A correctly designed process control is in effect continuously on the job, whereas a human operator cannot be, and would be a different individual depending on the pattern of work shifts. The Sanders system is thus designed to work unattended for extended periods of time, using a digital process controller and an electro-optic diameter sensor. Unattended operation has been achieved for periods over 100 hours.

Crystals growth by the Czochralski technique with a smooth outside surface have eliminated a source of defects. Manual control is difficult to provide because of long time constants (two to five hours) and long times (50 - 90 hours) required for successful growth. A number of diameter control techniques have been used, including weighing and infrared sensors. The weighing technique was considered difficult to implement in the Sanders furnace because of the relative masses and seal problems, and infrared sensors would not see the temperature gradient at the solid/liquid interface because fluoride melts are transparent in the near infrared (IR).

A system that detects the bright spot reflected from the meniscus between the crystal and melt, using a He-Ne laser source, and furnishes an analog signal proportional to the deviation from an optical null was designed to provide the necessary detection element. The source is modulated to distinguish its radiation from the background. The unit develops an analog output proportional to the deviation in diameter from the desired value.

The process is controlled and supervised by a digital computer, a Sanders MIP-16 high speed general purpose minicomputer. A digital processor provides long term stability and permits the programmed operations to be modified more readily than a hardwired analog processor. A significant feature is the large variety of logical responses that may be programmed to protect the crystal and the furnace from undersirable conditions. Such responses are particularly difficult to provide and modify in other equipment.

The program for the Sanders crystal growth system was adapted from a program prepared at MIT with modifications to make it compatible with Sanders equipment. The program consists of an executive section that schedules the tasks and provides housekeeping services, plus the functional programs that perform these tasks. The computer controls the furnace temperature, the position of the seed rod and crucible and a teletype. It receives input from a digital thermometer, the diameter sensor and the teletype. All operator interaction with the computer is made through the teletype using command mnemonics. A report is provided on an hourly basis that presents the current status of the system. All problems detected are typed immediately.

A special purpose interface was constructed to provide the interchange between the computer and the remainder of this apparatus. All input/output is handled by this interface except the teletype and paper tape reader.

#### 2.1.2.3 Diameter Sensor

The diameter sensor is mounted on the side of the growth furnace and uses one of the four viewing ports for optical access to the interior. A modulated helium-neon laser illuminates the meniscus between the crystal and the melt from which the crystal is growing. A bright spot of light is reflected from the meniscus that is related to the diameter of the crystal; this bright spot is observed by a dual-element optical detector. The output of the detector is processed to indicate the angular position of the spot within the field of view, and this information transferred to the computer.

The system is designed to operate in a "null seeking" mode that reduces the effect of dirty windows and other influences on signal strength. The signal is algebraically added to the temperature set points so that if the crystal is oversize, the temperature is incrementally increased to reduce the growth rate of the crystal; if the crystal is undersized, the temperature is decreased to obtain the opposite effect. The nature of the growth process requires a steady reduction in the furnace temperature as the crystal grows until the  $\text{LiF-LiYF}_4$  eutectic temperature is reached, at which point the growth process must cease.

### 2.1.3 FEED AND ATMOSPHERE PURIFICATION

Lasers for high peak power applications demand extreme purity of the host/activator combination. Important properties of these laser materials affected by impurities are susceptibility of the host crystal to radiation damage, light scattering from inclusions, and quenching of metastable lifetimes. To achieve the required improved purity levels, nearly all of the feed materials for crystal growth as well as virtually all of the auxiliary chemicals used in the processes must be purified, and laboratory equipment and air must be specially cleaned.

Purification and synthesis of fluoride materials are being carried out using a variety of modern wet chemical separation techniques, zone refining and a newly developed, low temperature hydrofluorination process. To obviate environmental recontamination during the chemical processing, all open operations are carried out in a Class 100 clean room. Closed equipment, including the centrifuge, carbon dioxide reactor, zone refiner, hydrofluorinators and fluoride growth furnace are located outside of the clean room. Recontamination during use of this equipment is normally not encountered.

The final step in feed purification, prior to use is treatment in a flowing hydrogen fluoride reactor. An inconel reactor is used to contain the HF, with a vitreous carbon boat for holding the material. The effluent from the reactor is trapped in a solution of potassium hydroxide (KOH), and the entire apparatus contained in a vented hood.

The growth of YLF crystals must take place under an atmosphere, rather than a vacuum, to reduce the evaporation of the melt. Two types of atmospheres can be considered: active and inert. An active atmosphere, such as HF, maintains a clean melt through its own chemical behavior. Other crystal growers have used such a gas, but there is a penalty because of its chemical activity toward many materials of construction, plus an added safety burden because of the large volume in the furnace chamber when compared to the hydrofluorination reactor. Crystals grown by the technique have exhibited both pump-induced and laser damage\*.

We have chosen to use extremely pure argon as an inert atmosphere. The gas is now available commercially with an impurity content of less than five parts per million (PPM) or can be purified by reaction with a heated charge of titanium that reacts strongly with almost all contaminants, and has a purity level of 1 PPM.

Figure 3 shows a flow chart of two alternate feed purification procedures which have been developed jointly by Sanders and MIT for assuring uniformity in feed purity. Feed purity is strongly related to resulting crystalline quality. These procedures have been shown to be effective in reproducing high optical quality material. Furthermore, these procedures permit the use of commercial oxide or fluoride starting materials. In the former case, selected impurities are eliminated by solvent extraction. The standard processing technique was used exclusively for this program.

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\*V. Rosati, ECOM (Private Communication)  
L. Esterowitz, NRL, (Private Communication)

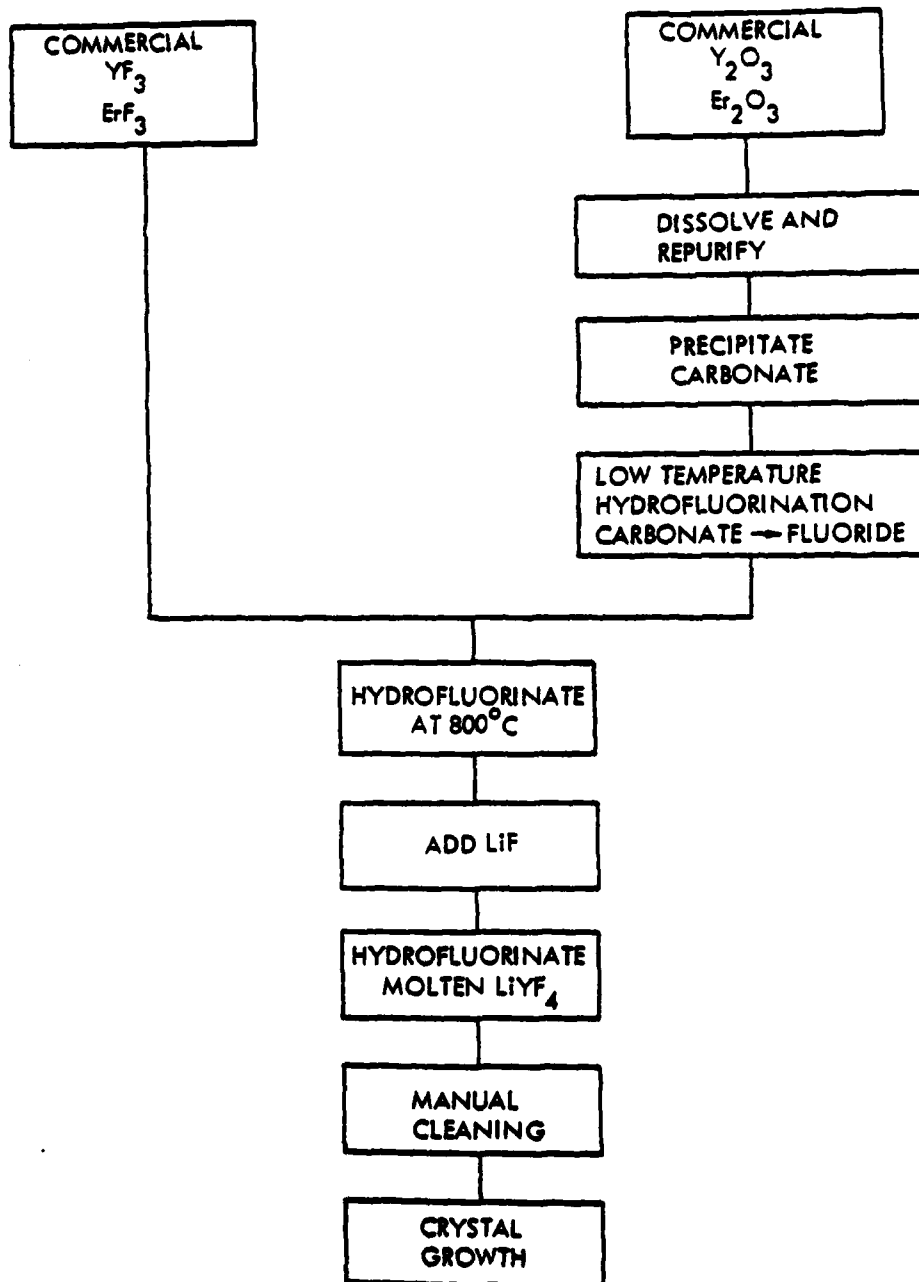


Figure 3. Feed Preparation Flow Chart

## 2.2 CRYSTAL GROWTH RESULTS

The results of the crystal growth processing and sample fabrication are presented in two summarizing tables. The growth summary (Table 1) describes material composition, growth parameters, and comments pertinent to the process and material evaluation. The fabrication summary (Table 2) list the laser samples by size and composition. This data is cross-referenced against sample identification number and boule number.

Eleven growth attempts were made during the period of this program. Eight boules of various composition were yielded. Two crystal furnaces were employed for this program. Initial difficulties with furnace 2 were solved early on in the program. Technical difficulties were attributed to a cracked flange allowing contamination of furnace atmosphere.

A major change in growth atmosphere was made during this period. Purity of argon was upgraded to a 99.9998% purity level. This change was made to further improve our process by reducing hydrocarbon content of the growth atmosphere. No further changes were made to the established growth process.

The quality of grown crystals was excellent. Very little scatter was observed. Laser samples were fabricated out of the highest clarity sections of boules. Although no laser damage measurements were made on these samples at Sanders, an internal IR&D program did address optical damage. Bulk damage testing was performed on YLF samples grown under a variety of conditions\*, the results were characterized by a wide scatter in the data, and no specific conclusions could be drawn. Under the passive testing, it appeared that pump energies ranging from 15-30 joules/cm<sup>2</sup> would be allowable without damage. The actual damage phenomena, as observed visually, seemed to be related to absorption at inclusion sites. Further characterization will be required to determine specific conclusions concerning damage resistance.

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\*Sanders Associates IR&D program NJG-80

TABLE 1. GROWTH SUMMARY

GROWTH RUN	COMPOSITION	GROWTH PARAMETERS	COMMENTS AND GROWTH EVALUATION
1	LiY.95Dy.05F <sub>4</sub>	F-2 Pull 1.5 mm/hr Atmosphere Purified Argon	Boule contained large concentration of scatter sites. Material was not regrown. A sample was fabricated and delivered.
2	LiY.98Ho.02F <sub>4</sub>	F-2 Pull 1.5 mm/hr Atmosphere purified Argon	First growth attempt resulted in low quality boule. Upon examination, a MgF <sub>2</sub> impurity was found in LiF chips. The second growth attempt yielded a high quality boule. Raw material inspection was instituted. Two samples were fabricated and delivered.
3	LiY.96Er.04F <sub>4</sub>	F-2 Pull 1.5 mm/hr Atmosphere - 99.9998 grade Argon	Satisfactory yield was obtained on first growth run. Low scatter was observed in material. One sample was fabricated and delivered.
4	LiY.92Er.08F <sub>4</sub>	F-1 Pull 1.5 mm/hr Atmosphere - 99.9998 grade Argon	Satisfactory yield was obtained on first growth run. Material displayed excellent optical quality. Two samples were fabricated and delivered.

TABLE 1. GROWTH SUMMARY (Continued)

GROWTH RUN	COMPOSITION	GROWTH PARAMETERS	COMMENTS AND GROWTH EVALUATION
5	LiY. <sub>96</sub> Ho. <sub>04</sub> F <sub>4</sub>	F-1 Pull 1.5 mm/hr Atmosphere 99.9998 grade Argon	First growth attempt was stopped due to poor crystal appearance (Polycrystalline). Second growth attempt yielded material of excellent optical quality. Two samples were fabricated and delivered.
6	LiY. <sub>84</sub> Ho. <sub>16</sub> F <sub>4</sub>	F-2 Pull 1.5 mm/hr Atmosphere - 99.9998 grade Argon	First growth attempt yielded sufficient material of good optical quality. Samples were fabricated and delivered.
7	LiY. <sub>84</sub> Er. <sub>16</sub> F <sub>4</sub>	F-2 Pull 1.5 mm/hr Atmosphere - 99.9998 grade Argon	First growth attempt failed due to power failure. Second attempt yielded high quality crystals. Samples were fabricated and delivered.
8	LiY. <sub>98</sub> r. <sub>02</sub> F <sub>4</sub>	F-1 Pull 1.5 mm/hr Atmosphere - 99.9998 grade Argon	First growth attempt yield sufficient material. Some scatter was observed. Samples were fabricated and delivered.

TABLE 2. SUMMARY OF LASER SAMPLE FABRICATION

GROWTH	BOULE	COMPOSITION	SAMPLE	
			IDENTIFICATION	SIZE
1	573	$\text{LiY}_{.95}\text{Dy}_{.05}\text{F}_4$	573.1	15 mm D x 5 mm L
2	583	$\text{LiY}_{.98}\text{Ho}_{.02}\text{F}_4$	583.1	5 mm x 5 mm x 30 mm
			583.2	5 mm x 5 mm x 20 mm
			583.3	4 mm D x 50 mm L
3	576	$\text{LiY}_{.96}\text{Er}_{.04}\text{F}_4$	576.1	5 mm x 5 mm x 10 mm
4	579	$\text{LiY}_{.92}\text{Er}_{.08}\text{F}_4$	579.1	5 mm x 5 mm x 10 mm
			579.2	5 mm x 5 mm x 20 mm
			579.3	5 mm x 5 mm x 30 mm
5	578	$\text{LiY}_{.96}\text{Ho}_{.04}\text{F}_4$	578.1	5 mm x 5 mm x 10 mm
			578.2	4 mm D x 50 mm L
6	585	$\text{LiY}_{.84}\text{Ho}_{.16}\text{F}_4$	585.1	5 mm x 5 mm x 10 mm
			585.2	4 mm D x 50 mm L
7	586	$\text{LiY}_{.84}\text{Er}_{.16}\text{F}_4$	586.1	5 mm x 5 mm x 10 mm
			586.2	4 mm D x 50 mm L
8	585a	$\text{LiY}_{.98}\text{Er}_{.12}\text{F}_4$	585a.1	4 mm D x 50 mm L
			585a.2	5 mm x 5 mm x 10 mm

A total of 16 laser specimens and rods were fabricated during this program. The fabrication specifications were for standard laser quality pieces. The following specifications were used for laser surfaces:

- Parallelism - 10 seconds of arc
- Power - /10
- Surface - 10/5 scratch/dig

Other surfaces were finished to a fine ground surface.

SECTION 3  
CONCLUSIONS AND RECOMMENDATIONS

3.1 CONCLUSIONS

Eleven growth attempts were made during this program, in which eight boules of various compositions were yielded. These boules were grown at two similar growth stations employing identical growth parameters. Crystal quality and yield improved in the later portion of the program, as the result of equipment upgrade and the switch to a high grade argon gas atmosphere.

A total of 16 samples and rods were fabricated and delivered to NRL under this contract. These samples were cut and polished to standard laser specifications.

3.2 RECOMMENDATIONS

Further study and development should address laser performance; i.e. damage resistance of the materials as a function of growth parameters and atmospheres. The majority of bulk and surface damage phenomena has been linked to extrinsic factors such as inclusions and bubbles. Growth parameter and atmosphere variations appear to strongly affect damage resistance of optical materials. A coherent program to evaluate these factors would increase the quality and the reliability of YLF laser materials.

